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Stimulated Rotational Raman Scattering in Nitrogen in Long Air Paths*

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Abstract

We studied the growth from amplified spontaneous emission of stimulated Raman scattering in air using a 20 cm diameter, linearly polarized, 1053 nm laser beam propagating over a 20-150 m air path. For 2.5 ns square pulses we find about 1% conversion on the S(8) and S(10) rotational Raman lines of nitrogen at an intensity-length product of 12 TW/cm, which implies a small signal gain coefficient of 2.5 cm/TW. For 1 ns square pulses, 1% conversion requires an intensity length product of about 16 TW/cm. The beam quality deteriorates severely above Raman threshold.

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Large fusion lasers propagate high intensity beams over distances of several tens of meters in air. Future larger fusion lasers will have even longer propagation paths at higher intensity and shorter wavelength. The propagation of high intensity beams through long air paths has also been proposed for other purposes. It is therefore important to understand the onset of nonlinear processes such as stimulated Raman scattering in long air paths so that degradation of beam quality due to stimulated scattering can be avoided. Averbakh, Makarov, and Talanov[1] show that for pressures below about two atmospheres in air the process of lowest threshold is stimulated rotational Raman scattering (SRRS) in nitrogen. This is confirmed by other unpublished calculations.[2] We have conducted an experiment using the LLNL Nova fusion laser[3] to verify this conclusion and to assess the impact of SRRS on large fusion laser systems.

We diverted a section of one of the ten Nova laser beams during the initial activation and test of the laser to study the growth from amplified spontaneous emission of SRRS in the nitrogen in long air paths. The 1053 nm test beam was 20 cm in diameter, collimated, and linearly polarized. We determined the average intensity in the test beam using energy, pulselength, and beam profile measurements which are integrated into the laser system.[3] We studied both a well-polarized section of uniform intensity near the center of the 74 cm diameter Nova beam and a somewhat less uniform section with 5% of its energy in the orthogonal polarization near the edge of the beam. The test beam propagated in air in the Nova high bay over a path length of 20-150 meters. Path lengths longer than 85 meters used a high reflector to fold the beam path once. At the end of the propagation path we diffused a 2.5 cm sample from the center of the beam and photographed its spectrum using a film spectrograph. We estimate that about 10^{-2} photon conversion to the first Stokes is detectable in this arrangement and define "threshold" as any observable film exposure at the first Stokes wavelength. We also photographed the intensity distribution in the beam before and after the propagation path.

Figure 1 shows the SRRS components we observe as a function of the average intensity times propagation distance $I_{pa}L$, for several distances and two pulse lengths. For a 1 ns full width at half maximum (FWHM) flat top ($\sim$ 0.1 ns rise and fall) pulse we begin to see first Stokes components of nitrogen SRRS at $I_{pa}L$ of 12 TW/cm at the edge of the Nova Beam and 16 TW/cm in the center of the beam. The edge of the beam has more intensity modulation than the center and we estimate that the 1±0.33 intensity modulation is consistent with a peak intensity-length product of 16 TW/cm, which agrees with the threshold in the center of the beam where the modulation is much smaller. With a flat top 2.5 ns FWHM pulse we observe a threshold of 12 TW/cm in the clean central part of the beam. The data are consistent with the expected γI_pL dependence where γ is a Raman gain coefficient. Diffractive and nonlinear index effects change the details of the beam intensity profile over the propagation path so it is difficult to be more quantitative about intensity-length products. The final spatial filter[3] forms an image of the last laser amplifier at the 65 meter point in the propagation path, so some diffractive effects are suppressed by image relaying.

Figure 2 shows a characteristic spectrum of nitrogen SRRS, well above threshold, at the point indicated by a dashed circle on Figure 1. This spectrum was recorded on Kodak Tri-X film, which was digitized and linearized by comparing film density to the density induced by a series of 1.053 nm, 1 ns exposures of known intensity ratio. Tri-X film is insensitive to normal exposure at wavelengths longer than 700 nm, but short high-intensity pulses will cause an exposure and this technique is used routinely at this laboratory for semi-quantitative intensity diagnostics. The spectrum shows significant first Stokes intensities on the S(6), S(8), and S(10) rotational Raman lines at 59, 75, and 91 cm^{-1} shift from the laser frequency. There are numerous second and third Stokes components corresponding to various intercombinations of these first Stokes components. Weak fourth Stokes and first anti-Stokes components were observed on some shots. We estimate that for this spectrum 62% of the original pump energy remains in the pump with 20%, 15%, and 3% of the energy shifted to first, second, and third Stokes components, respectively. Although the S(6) line is the most intense in the figure, we find that the S(8) and S(10) lines are the first to go over threshold and that they do so at the same value of I_pL to within the precision of this experiment. Rotational redistribution and four-wave mixing effects far above threshold lead to a very complex spectrum, as previously noted by Averbakh.[1] Photographs of the beam well above Raman threshold show that the Raman components have a speckle pattern with a characteristic scale of order 1-3 mm and are highly divergent, occupying an area two to three times that of the collimated pump beam at the end of the propagation path. The intensity uniformity of the remaining pump also degrades severely.

The pulse lengths used in this experiment are short enough that transient effects must be considered in the buildup of amplified spontaneous emission. The nitrogen rotational Raman lines have a linewidth (FWHM) of about 2.4 GHz at one atmosphere[4], which corresponds to a relaxation time τ_0 of 0.133 ns. Analyses[5,6] of transient Raman scattering show that pulses longer than about 20 τ_0 show only a very minor pulselength dependence in the Raman gain required for amplified spontaneous emission to grow to a significant intensity. Shorter pulses require a larger value of $\gamma I_p L$ to show significant growth. Our data at 2.5 ns are at about 20 τ_0 for nitrogen so we shall consider them to be representative of steady-state Raman scattering though longer pulses at the same intensity should have a Raman threshold perhaps five or ten percent lower. Our 1 ns (8 τ_0) results show that $I_p L$ must be about 30-40% higher than at 2.5 ns to reach the same Raman conversion. This is consistent with typical models of transient scattering.[5,6]

It is well-established that Raman small signal gains of order 20-30 Nepers (e^{20-30}) are required for stimulated Raman processes to grow to significant conversion by amplified spontaneous emission under steady state conditions, but it is rare to see these estimates explicitly displayed though they are easily calculated using well-known principles. We shall therefore do that here to illustrate the dependence of "threshold" on conditions since a weak scatterer with a long wavelength pump requires significantly higher gain to reach threshold than a strong scatterer with a short wavelength pump, and this point can be missed.

Consider a long cylindrical Raman gain region defined by the pump beam aperture and propagation path. The intensity $I_s(\nu)$ of the spontaneous Stokes emission from a length L into solid angle $d\Omega$ and bandwidth $d\nu$ is[7,8]

$$I_s(\nu) = I_p \; N_\ell L \frac{d\sigma}{\partial\Omega} f(\nu) \; d\nu \; d\Omega \tag{1}$$

where $\partial\sigma/\partial\Omega$ is the differential Raman scattering coefficient per molecule integrated over the fluorescence lineshape $f(\nu)$, and N_ℓ is the number of molecules in the lower level per unit volume. We assume $\partial\sigma/\partial\Omega$ independent of angle and pump and Stokes beams of a single distinct polarization. The Raman intensity gain over the length L is exp $\{G(\nu)\}$ where[7,8]

$$G(\nu) = \gamma(\nu) I_p L = \frac{\lambda_s^2}{h\nu_s} \left(N_\ell - \frac{g_\ell}{g_u} N_u\right) \frac{\partial\sigma}{\partial\Omega} f(\nu) \; I_p L, \tag{2}$$

N_u is the density of molecules in the upper level, and g_ℓ, g_u are degeneracies.

Substituting (2) in (1) at the center of the line ($\nu = \nu_0$) and assuming a Boltzmann distribution gives

$$I_s(\nu_0) = \gamma(\nu_0)\ I_p L \frac{h\nu_s}{\lambda_s^2} \left(\frac{1}{1-e\left(-\frac{h\nu_R}{kT}\right)}\right) d\nu\ d\Omega \tag{3}$$

As input noise it is reasonable to take the spontaneous emission from the first gain length of the amplifier ($\gamma(\nu_0) I_p L = 1$) since emission from other regions of the amplifier is at least a factor of e less important. It can be shown that this is equivalent to assuming a noise input of one photon per $1/d\nu$ seconds in each mode of the radiation field within the solid angle $d\Omega$ defined by the gain region.[9] The bandwidth $d\nu$ is the region over which the amplifier gain is large enough that amplified spontaneous emission within that bandwidth makes an important contribution to the output. Using a mildly circular argument, we know that a gain G of order 25 Nepers will be required. If G = 25 changes by 3%, e^G the small signal gain changes by a factor of two, which defines the half-power points of the output spectrum and is a reasonable approximation for $d\nu$. If $f(\nu)$ is roughly Gaussian then $d\nu \sim 0.2\ \Delta\nu$ where $\Delta\nu$ is the FWHM of the Raman line.

The Stokes intensity will grow exponentially with G until it becomes a major fraction of the pump intensity I_p. If the Stokes output intensity under small signal conditions is $(\nu_s/\nu_p) F I_p$ then "threshold" by our previous definition is at F = 0.01 and hard saturation (50% photon conversion in a saturation model[7,8] in the absence of other limiting effects) is at F = 1. The small signal gain $G = \gamma I_0 L$ required to reach a particular value of F is

$$G = \ln F + \ln \frac{\nu_s}{\nu_p} + \ln \frac{5\lambda_s^2 I_p}{h\nu_s d\Omega\, \Delta\nu} + \ln \left(1 - e^{-\frac{h\nu_R}{kT}}\right) \tag{4}$$

Setting F = 0.01, $d\Omega = 3 \times 10^{-6}$ steradians for a 20 cm beam propagating 100 m, and using other parameters previously defined, we have G = 32 Nepers for the Stokes intensity to reach "threshold" in the present experiment for steady-state scattering.

The input noise intensity could come from a source other than the first gain length of the propagation path. In this experiment, for example, the Nova disk amplifiers are filled with nitrogen gas so there is some amplified Stokes fluorescence from within the laser itself. The laser output beam goes through a spatial filter before the propagation path used in this experiment and that spatial filter passes a solid angle of about 3×10^{-7} steradians or much less than the 3×10^{-6} steradians subtended by the propagation path, but if there were more than 2.3 Nepers of gain in the nitrogen in the laser chain, then the noise input from within the laser would exceed the noise from the first gain length of the propagation path into its larger solid angle. We estimate that the SRRS gain within the laser chain could be as high as 4-6 Nepers under the highest laser output conditions so the noise input from within the laser is significant and probably reduces the gain required to reach threshold by two or three Nepers, which is a small change given the accuracy of the analysis and the intensity measurements. Note also that grazing incidence reflection from the walls of a beam pipe could increase $d\Omega$.

The 2.5 ns data of Fig. 2 show that an I_pL product of 12 TW/cm gives adequate gain to reach threshold and we argue above that this requires a gain of about 30 Nepers in the air path (32 Nepers total). This implies a Raman gain coefficient γ for the rotational Raman lines of nitrogen in air which is about 2.5 cm/TW. Averbakh, et al.,[1] calculate a steady-state Raman gain coefficient of 6.6 cm/TW for the S(8) and S(10) transitions in pure nitrogen excited by a circularly polarized beam at 532 nm. The gain in our experiment is lower by factors of 0.5 due to pump frequency, 0.67 due to linear rather than circular polarization[1], and 0.8 due to the concentration of nitrogen in air, so Averbakh's calculation predicts a steady-state gain of 1.8 cm/TW for our conditions and our inferred value is higher than this calculation by a factor of about 1.4. Averbakh's threshold measurements using 1 ns pump pulses are nominally in agreement with his calculated gain so we infer that his experiment supports the conclusion that the gain for 2.5 ns pulses is higher than his calculated value by a factor of about 1.4 as we see in our pulse length dependence. Averbakh shows transient effects for pulses less than 1 ns but does not discuss longer pulses. We conclude that our inferred gain is in agreement with Averbakh's measurement and that the absolute accuracy is a few tens of percent.

Bloembergen[10] has predicted that the coupling between Stokes and anti-Stokes waves can suppress Raman gain when the conversion of two pump photons to a Stokes plus anti-Stokes photon is exactly phase matched. Averbakh,[1] and Perry,[11] see effects which they attribute to this process. In a gas with very low dispersion and small Raman shift, such as in our present experiment, the phase-matched direction is close to exact forward propagation and one might expect a suppression of forward gain, but we do not see any such suppression. The gain suppression requires very precise phase matching, so Stokes waves propagating at an extremely small angle with respect to the exact phase matched direction will see the normal high gain. Our gain region has a large Fresnel number and the output Stokes beam has a much higher divergence than the pump so a wide range of angles is indeed present. It is possible that there is no Stokes radiation in a very narrow angular range around the exact phase-matched direction, but if the suppression occurs, it does so over such a small angular range that it is unimportant in geometries with a large Fresnel number.

We conclude that the gain for stimulated rotational Raman scattering in nitrogen in air at 1053 nm for 2.5 ns pulses is 2.5 cm/TW to an accuracy of a few tens of percent. Since 2.5 ns pulses are still mildly transient, the gain for true steady-state pulses is higher by $\sim$ 5-10%. In large glass laser systems deterioration of the beam quality due to stimulated Raman scattering will begin when the peak intensity-length product in air is about 10-12 TW/cm for long pulses or 16 TW/cm for 1 ns pulses, with a further increase for shorter, more transient pulses. Serious degradation will require 20-30% higher I_pL. Normal operating conditions of the Nova laser are below this level, though there will be some degradation of the beam at maximum output ($\sim$ 9 kJ/beam in 3500 cm^2 at 1 ns, 65 m propagation path). Longer beam paths with more intense beams will require that the air be replaced by a gas with lower Raman cross-section.

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References

1. V. S. Averbakh, A. I. Makarov, and V. I. Talanov, Sov. J. Quantum Electron., 8, 472-476 (1978)
2. G. C. Herring, M. J. Dyer, and W. K. Bischel, submitted to Optics Letters, and J. Jacob, Science Research Corporation, and N. A. Kurnit, Los Alamos National Laboratory, private communications.
3. W. W. Simmons and R. O. Godwin, J. Nucl. Tech/Fusion, 4, 8 (1983); W. W. Simmons, et al., Proc. of the 9th Symposium on Engineering Problems of Fusion Research (Chicago, 1981) pp 1221-1273, IEEE publication #81CH1715--1 NPS.
4. K. S. Jammu, C. E. St.John, and H. L. Welsh, Can. J. Phys, 44, 797-814 (1966).
5. R. L. Carman, F. Shimizu, C. S. Wang, and N. Bloembergen, Phys. Rev., A2, 60-72 (1970).
6. A. Laubereau and W. Kaiser, Rev. Mod. Phys, 50, 607-665 (1978).
7. W. Kaiser and M. Maier, Chapter E2 of Laser Handbook, vol 2, F.T. Arrechi and E. O. Schulz-Dubois, eds., North Holland, Amsterdam, 1972.
8. J. R. Murray, J. Goldhar, D. Eimerl, and A. Szoke, IEEE J. Quantum Electron., QE-15, 342-368 (1979).
9. See chapter 24 and appendix 5 of A. Yariv, Quantum Electronics, Wiley, NY, 1967.
10. Section 4-6 of N. Bloembergen, Nonlinear Optics, Benjamin, NY, 1965.
11. B. N. Perry, P. Rabinowitz, and D. S. Bomse, Opt. Lett. 10, 146-148 (1985).

Figure Captions

1. Stimulated Raman Stokes and anti-Stokes orders observed for laser shots characterized by the product of average intensity and propagation distance in TW/cm.
2. Spectrum of rotational stimulated Raman scattering from nitrogen in a 150 m air path at an intensity-path length product of 30 TW/cm for a 1 nsec laser pulse.

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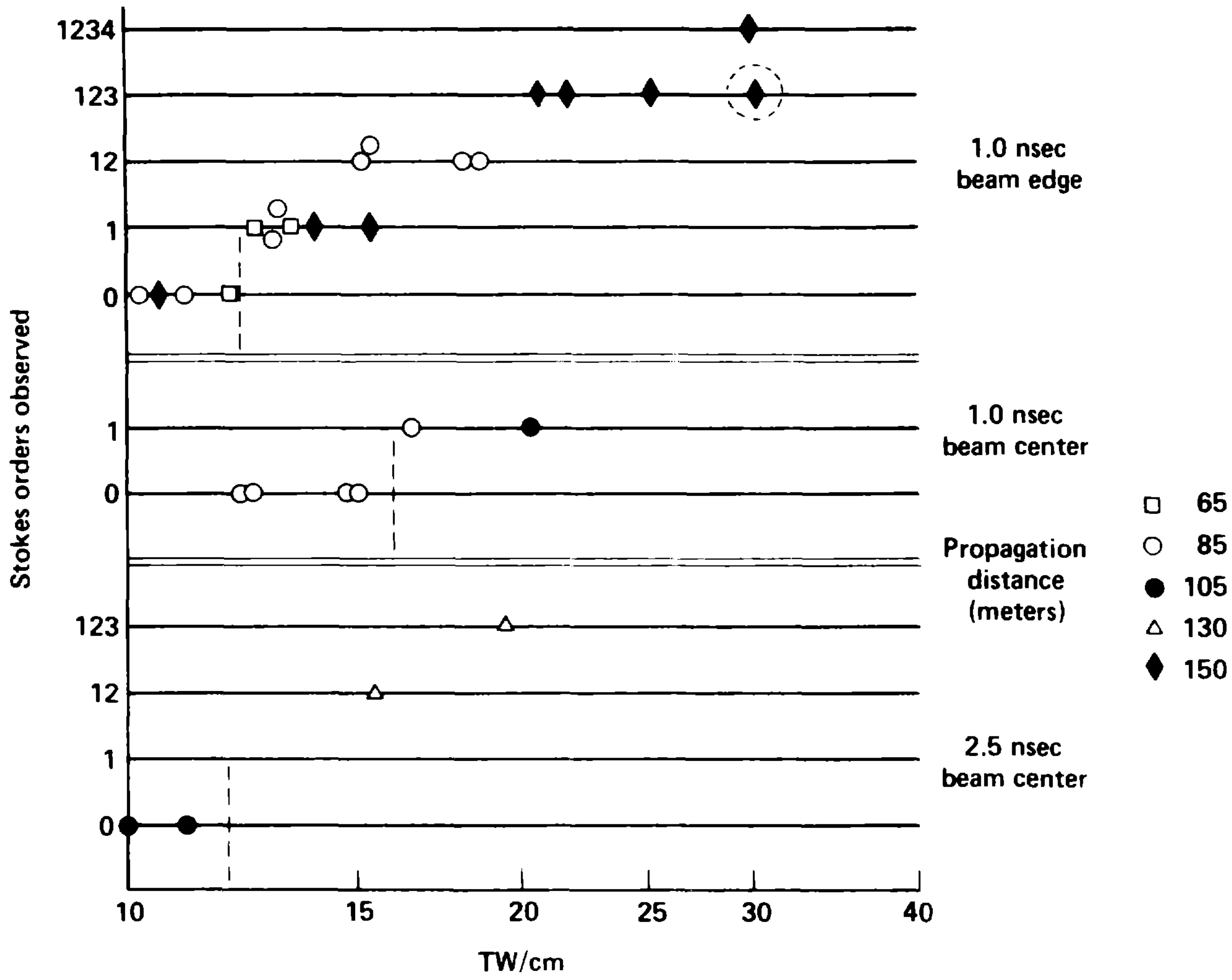
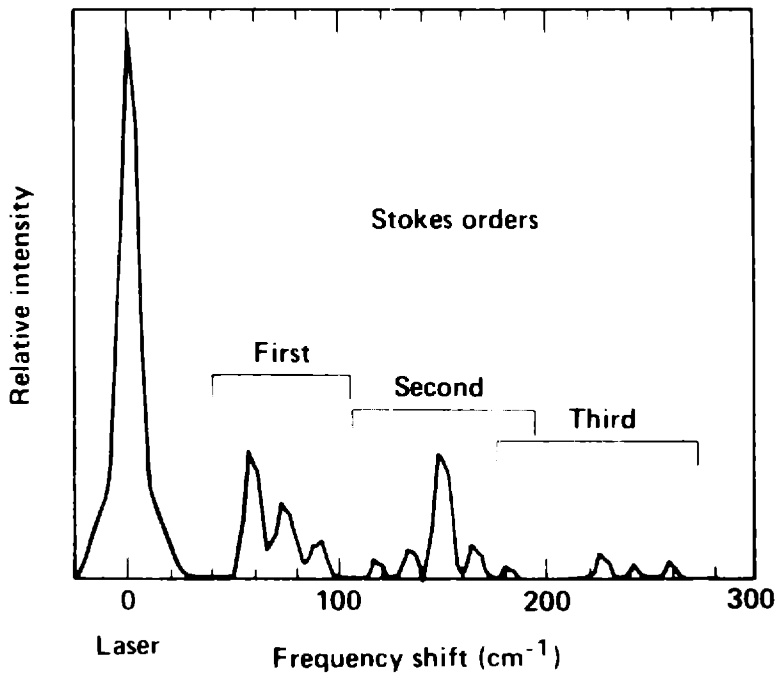

1234
123
12
1
0
1
0
123
12
1
0
Stokes orders observed
1.0 nsec
beam edge
1.0 nsec
beam center
Propagation
distance
(meters)
2.5 nsec
beam center
65
85
105
130
150
10
15
20
25
30
40
TW/cm

Figure 1



Relative intensity
Stokes orders
First
Second
Third
0
100
200
300
Laser
Frequency shift (cm-1)

Figure 2